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Developing Biopolymer Based Edible Films with Improved Anti-Microbial Properties †

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Abstract: The food packaging industry is projected to reach a worth of $423.27 billion by 2025. Due to the hazardous impact of synthetic polymers on the environment and human and animal health, there is an increasing focus on biopolymer-based edible and biodegradable food packaging films. This study aimed to develop composite films made from polysaccharide (pectin) and protein (gluten) with castor oil as a hydrophobic agent to enhance moisture barrier properties. A statistical 22 factorial design of experiments was employed, with gluten and castor oil percentage chosen as the two factors. All films were evaluated for sensory, moisture barrier, mechanical, surface hydrophobicity, morphological, and biodegradability properties. Films made from 10% *w/w* gluten and 15% *w/w* castor oil demonstrated the best moisture barrier and tensile properties. Addition of castor oil enhanced the hydrophobicity and reduced moisture permeability by two times compared to control pectin films.

Keywords: biodegradable films; biopolymers; coatings; edible films; films food packaging; pectin; castor oil; gluten; water vapor transmission rate; contact angle; tensile strength; anti-microbial

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1. Introduction

The increasing concern for environmental sustainability has led to the development of biodegradable packaging as an alternative to conventional plastics. In this context, the current project focuses on developing edible and biodegradable films using pectin and gluten biopolymer for food packaging applications. The film's properties, such as transparency, moisture content, thickness, moisture barrier properties, mechanical and thermal stability, depend on various process parameters. This study aims to optimize the process parameters, such as concentration of film-forming solution, concentration of gluten, relative concentration of plasticizer, temperature, and humidity for drying. The results suggest that the optimum process parameters are 10% gluten for 2.5 *w/v*% of film-forming solution with 15% castor oil, at a temperature of 45 °C and a humidity of 60%. The optimized films exhibit less thickness, high moisture barrier, high transparency, mechanical stability (tensile strength), and flexibility compared to control pectin films. This research could contribute to sustainable food packaging practices with biodegradable films.

2. Materials and Methods

2.1. Materials

Chemical grade extra pure pectin with a molecular weight ranging between 30,000 to 100,000 was purchased from Loba Chemie Pvt. Ltd., Mumbai, India. The degree of esterification (DE) of the pectin was 63–66%, while the methoxyl content was between 6–10%.

2.2. Methodology to Develop Films

To prepare the control pectin films with a weight/volume (*w*/*v*) of 2.5%, 2.5 g of pectin powder was added to 100 mL of distilled water, and the solution was homogenized for 25 min at a constant temperature of 45 °C and 600 rpm using a magnetic stirrer with heating arrangement. The larger dispersed particles were subdivided by homogenizing the solution with a homogenizer (IKA T25 ULTRA-TURRAX) at 3000–5000 rpm after the solution had cooled to room temperature. The pH of the resulting solution was measured with a pH meter and adjusted by adding sodium hydroxide buffer tablets. To agitate the smaller particles, the solution was kept in an ultrasonic homogenizer for 45 min. Then, 40 mL of the hot solution was poured into a 100 mm diameter petri dish to form films. The films were dried in a humidity chamber (NECSTAR NEC-HTC-150) under controlled temperature and relative humidity conditions. After drying, the films were peeled off and stored in a vacuum desiccator. The same process was used to prepare all the films based on the statistical design of experiments, which included castor oil as a plasticizer and gluten as part of the film-forming solution.

2.3. Characterization

2.3.1. Moisture Content

To determine the bound moisture content of the film, a 3 cm × 3 cm film was placed in a hot air oven at a temperature of 100 ± 5 °C for 3 h. The percentage weight loss of the film compared to its initial weight was then calculated using $(W_{\text{initial}} - W_{\text{final}}) \times 100/W_{\text{initial}}$. Where, W_{initial} is the initial dry weight of the sample film, and W_{final} is the final dry weight of the sample film.

2.3.2. Solubility

The water solubility of the films was tested by soaking a 3 cm × 3 cm film in 15 mL of water. Time for complete dissolution of films was noted.

2.3.3. Thickness

To measure the thickness of the film, a micrometer screw gauge with a least-count of 0.001 mm was used. the thickness at five random positions on the 100 mm diameter film was measured and the average of the values was reported as the film thickness.

2.3.4. Transparency

To perform the transparency test using the Spectrophotometer (CHN SPEC & CS-580A), a transparent film was first placed on a white plate, and the colorimeter is calibrated using the standard values of the transparent film ($L^* = 86.75$, $a^* = 0.93$, and $b^* = 1.03$). The film to be tested is then placed on the white plate, and the colorimeter is used to measure the L, a, and b values of the film. These values are then used to calculate the total color difference/transparency parameter (E) and whiteness index (WI) using the formulas shown below:

$$\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2} \tag{1}$$

$$WI = 100 - \sqrt{(100 - L)^2 + a^2 + b^2} \tag{2}$$

2.3.5. Water Vapor Transmission Rate

The Water Vapour Transmission Rate (WVTR) analysis of the films was conducted at the Northern India Textile Research Association (NITRA), Ghaziabad, India, in accordance with ASTM standard ASTM E96/E96M-05 (water method), maintained at a temperature of 32 ± 2 °C and a relative humidity of 50 ± 2%, with air velocity set between 0.02–0.3 m/s. The WVTR of the samples in the present study is reported in units of g/m^2/day. The analysis was done by using the facility at Northern India Textile Research Association, Ghaziabad, India and Sree Chitra Tirunal Institute for Medical Sciences & Technology, Thiruvananthapuram, India.

2.3.6. Mechanical Properties

Mechanical properties of the films were studied using ASTM D882 method. A standard specimen of dimensions 10 mm width and 250 mm length was used for the analysis. A crosshead speed of 10 mm/min was used. The analysis was done by using the facility at Sree Chitra Tirunal Institute For Medical Sciences & Technology, Thiruvananthapuram, India.

2.3.7. Water Contact Angle

The water contact angle (WCA) of the films was measured at room temperature using sessile drop method by using a video based contact angle measuring device (Data Physics OCA 15 Plus, Germany) and imaging software (SCA20 software, Germany) within 10 s after introduction of water droplet. Six independent measurements at different sites on the sample are performed for the contact angle study. The analysis was done by using the facility at Sree Chitra Tirunal Institute For Medical Sciences & Technology, Thiruvananthapuram, India.

2.3.8. Fourier Transformation Infrared (FTIR)

FTIR analysis of the films was carried out using a Fourier transform infrared spectrophotometer. The spectrum of each film was obtained in the range of 4000–500 cm^{-1} with a resolution of 4 cm^{-1}. The results obtained from the FTIR analysis provided insights into the chemical composition and structural features of the films.

2.3.9. Anti-Microbial Test

The *E. coli* strain was uniformly spread on agar-agar plate. Clove oil was added to the film solution to test for antimicrobial property using the growth of inhibition.

2.3.10. Biodegradability and Shelf Life Test

The standard procedure was followed to prepare the film for the biodegradability test, and it was monitored continuously after being sown in soil. Once the film was biodegraded, some seeds were planted in the same spot to ensure the soil quality. To coat the fruits, film solution was applied, and the capsicum, bananas, and chillies were repeatedly dipped until an even coating was achieved.

3. Results and Discussions

3.1. Design of Experiment

All the films developed were easy to peel off from the petri dish and were flexible enough. The films were stored in vacuum desiccator to keep away from moisture contamination. The amount of gluten and castor oil added as per the design of experiments is mentioned in the Table 1.

Table 1. Details of low and high levels of the gluten and castor oil.

Run No.	Gluten (g)	Castor Oil (g)
1	0.75 (+1)	0.125
2	0.25 (−1)	0.125 (−1)
3	0.75	0.375 (+1)
4	0.25	0.375

3.1.1. Moisture, Water Solubility, Thickness and Transparency

The biopolymer films developed in the present work had a bound moisture content of 0.04% to 0.1%. The films were completely soluble in water due to the presence of hydroxyl and carboxyl groups. The thickness of the films was observed to be around 0.125 ± 0.004 mm. All films had good transparency, with the transparency parameter ΔE value ranging from 11–25 and WI ranging in 82–85.

3.1.2. Water Vapor Transmission Rate (WVTR)

The pectin and gluten films integrated with castor oil developed in the present work demonstrated excellent moisture barrier properties. The WVTR of the films was observed to be in the range of 1295–2082.1 gm^{-2}day^{-1}. A comparison graph is presented in Figure 1. Figure 1 shows that the gluten and pectin composite film exhibited high WVTR. These films contain 30% gluten. As both pectin and gluten are moisture sensitive, the moisture transmission rate was higher. Comparing the WVTR of the films developed as shown in Figure 1, it is evident that the addition of oil to pectin reduced the WVTR. This indicates the increase in moisture barrier of pectin film with the incorporation of castor oil. Comparing the WVTR of pectin + gluten films with that of "pectin + gluten + castor oil", it is evident that the moisture barrier properties significantly improved with the incorporation of castor oil.

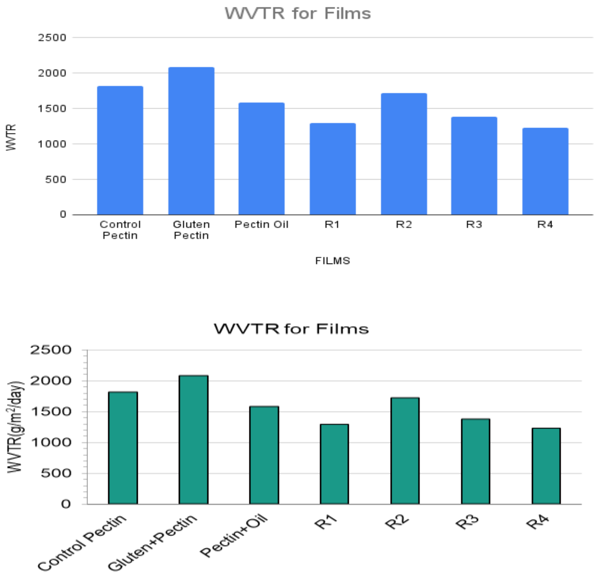

Figure 1. Comparison of water vapor transmission rate of the pectin + gluten + castor oil films.

3.1.3. Water Contact Angle

The contact angle measures wettability and higher contact angles indicate higher hydrophobicity. The water contact angle of the films after 10 s of sessile drop are presented and compared in Figure 2. The Figure 2 shows that R1 and R4 films had the highest contact angles of 76.7° and 78°, indicating higher hydrophobicity, while R2 had the lowest contact angle of 53.3°, indicating lower hydrophobicity. Control and gluten pectin films had similar contact angles of 61.4° and 60.4°, respectively, while pectin, castor oil had a slightly higher contact angle of 71.4°. The addition of castor oil to the pectin and pectin + gluten films could enhance their surface hydrophobicity.

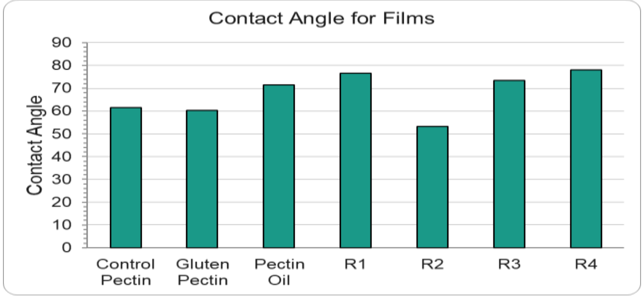

Figure 2. Comparison of water contact angle of the pectin + gluten + castor oil films.

3.1.4. Mechanical Properties

The mechanical properties such as tensile strength, load at break and percent strain at break are reported and compared in Figure 3. Figure 3 suggests that the film R2 had the highest tensile strength and R4 the maximum load at break, indicating that lower gluten content with higher plasticizer content may improve mechanical properties. R3 had the lowest tensile strength and load at break, suggesting that higher plasticizer content may not always improve mechanical properties. Overall, these findings suggest that optimizing the gluten and plasticizer content can lead to films with improved mechanical properties.

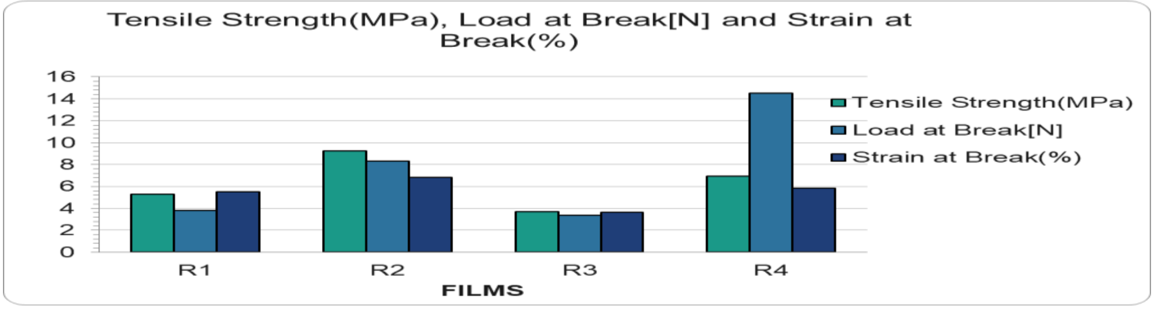

Figure 3. Comparison of mechanical properties of the pectin + gluten + castor oil films.

3.1.5. Fourier Transform Infrared (FTIR)

The FTIR spectra of the "pectin + gluten + castor oil" films when compared with the FTIR of pure pectin, gluten and castor oil suggest that the mixing of the three components is pure physical and no chemical alteration happened during processing. However, there was a shift in peak intensity and position. Peaks at 3300 cm^{-1} were associated with O-H stretching vibrations of hydroxyl groups in pectin, and peaks at 1650 cm^{-1} corresponded to the C=O stretching vibration of the carbonyl group in pectin. Peaks at 2920 cm^{-1} and 2850 cm^{-1} corresponded to C-H stretching vibrations of aliphatic chains present in castor oil and gluten.

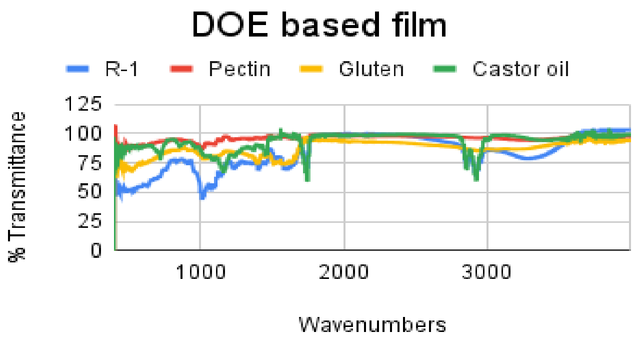

Figure 4. FTIR of the pectin + gluten + castor oil film.

3.1.6. Biodegradability and Shelf Life Test

The films got completely degraded within 24 h and had no negative impact on soil quality. As shown in Figure 5, they were also effective in preventing spoilage of food products, such as capsicum, bananas, and green chillies, resulting in longer shelf-life and fewer black spots. The film coating can enhance the shelf-life of food products and prevent spoilage effectively.

Figure 5. Shelf life improvement properties of pectin + gluten + castor oil films.

3.1.7. Anti-Microbial

The zone of inhibition formed was 3.2 cm thereby indicating the effectiveness of the film solution with clove oil as an antimicrobial agent as shown in Figure 6.

Figure 6. Antimicrobial properties of pectin + gluten + castor oil films with clove oil.

4. Conclusions

The present work reports biodegradable films developed from pectin and gluten biopolymers for food packaging applications. Castor oil was used as hydrophobic additive. The films exhibited promising properties such as thinness, high transparency, good solubility, low moisture content, and enhanced moisture barrier and mechanical properties. The films were also found to be biodegradable and did not affect soil quality. Antimicrobial tests showed that pectin-based films with clove oil have potential as an antimicrobial agent. The pectin-based films with gluten and castor oil demonstrated improved mechanical and water vapor barrier properties compared to control pectin films. The results suggest that pectin-based films with gluten and castor oil have promising potential as edible packaging materials with enhanced mechanical, barrier, and antimicrobial properties. Further studies are required to investigate their performance in real-world packaging applications. This research could contribute to sustainable food packaging practices with biodegradable films.

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Conflicts of Interest: The authors affirm that they do not have any known financial interests or personal relationships that could have influenced the reported work in this paper.

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